Heterogeneities and local fluctuations in glassy systems

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ABSTRACT

The aim of this report is to review a theoretical approach that has been proposed recently to describe dynamic fluctuations in glassy systems (work in collaboration with H. E. Castillo, C. Chamon, P. Charbonneau, J. L. Iguain, M. P. Kennett, D. R. Reichman and M. Sellitto). Firstly, I summarize some of the main features of the averaged and global non-equilibrium relaxation of glassy systems, weakly sheared viscous liquids and weakly tapped granular matter, and how these results have been successfully reproduced with a mean-field-like analytic approach. Secondly, I explain the outcome of more refined experimental and numerical measurements that point at examining the dynamics at a mesoscopic scale, and the role of noise measurements in this respect. Finally, I discuss how our theoretical approach can be confronted to new experimental and numerical tests.

Keywords: Glassy systems, non-equilibrium dynamics, spatial and temporal fluctuations

1. INTRODUCTION

A system may not be able to reach equilibrium with its environment for several reasons.[1] The most common cases are those in which the time needed to equilibrate the sample falls beyond the experimental time-window. Examples are given by domain growth, phase separation and classical and quantum glassy systems. On the other hand, external forces can drive a sample out of equilibrium, as in rheological measurements. If the systems tested have a tendency to evolve slowly, and if the perturbations used are not too strong, their slow dynamic behaviour may resemble strongly the one of glasses. This is the case, for instance, of sheared super-cooled liquids, and weakly tapped granular matter.

Even if there is a kind of universality in the form of the decay of bulk observables in glassy systems, its origin might vary significantly from material to material. In some systems it is pretty easy to identify the mechanism behind the dynamic arrest and the slow non-equilibrium dynamics at low temperatures. For instance, a ferromagnetic spin model with quenched random fields has a very slow coarsening dynamics since the interfaces get pinned by disorder. In more generic glassy materials, the impressive slowing down demonstrated by the growth of the relaxation time by several orders of magnitude when approaching the glass transition is not accompanied by any detectable change in structure. Indeed, establishing a relation between dynamics and structure in glassy materials has remained elusive.

The recent development of powerful experimental techniques[2–10] and the use of extensive numerical simulations[11–22] are starting to yield very interesting information about the dynamic behavior at a mesoscopic scale. In these studies, emphasis is set on identifying the most relevant rearrangements that take place during evolution. In addition, a detailed analysis of temporal fluctuations in the evolution of the relevant observables yields complementary information that allows one to distinguish between intermittent and continuous dynamics, the former being associated with sudden, large rearrangements.[6, 9, 10] The insight obtained from these studies might be very valuable to understand the reason for the glassy arrest and to develop a complete theoretical description of it. The examples cited in the first paragraph belong to a particular class of out of equilibrium systems: those with a *very slow dynamics*.[1] Exploiting this property, as well as other more subtle ones,[23–28] one can hope to develop a common theoretical description of all of them. Indeed, the decay of different averaged two-time global correlation functions in rather diverse glasses have a very similar pattern. For example, the averaged two-time correlations

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between global magnetic fluctuations in disordered spin systems,[29] and the averaged two-time intensity-intensity correlations in light scattering experiments in colloidal suspensions decay with a very similar form,[10, 30–32] see Fig. 1. In the paramagnetic or super-cooled liquid (stationary) and the spin-glass or jammed (aging) phases these are rather well described analytically by the solution to the dynamics of fully-connected disordered spin models.[23, 24] In the late 80s Kirkpatrick, Thirumalai and Wolynes[23] showed that, above the glass transition, these models lead to dynamic equations for the disorder and statistical averaged two-time functions that are identical to those derived within the mode-coupling approach to super-cooled liquids.[33] More recently it has been realized that these models extend the mode-coupling approach below the glass transition.[26, 34, 35] The precursor two-step relaxation of averaged bulk correlations in super-cooled liquids, the growth of the structural relaxation time when lowering the external temperature, aging effects in the glassy phase, shear thinning effects, etc. are captured by these models.[1]

One of the more interesting and open theoretical problems now is how to include in such a description large-scale fluctuations (in space and time) that are possibly associated to cooperative and correlated rearrangements of particles.

In equilibrium and away from criticality any global observable of a macroscopic system has Gaussian fluctuations. At criticality, instead, one observes non-Gaussian fluctuations due to the divergence of the correlation length and the non applicability of the central limit theorem. Still, scale invariance at the critical point constrains the possible probability distributions; these are determined by the universality class to which the systems belong. A similar criterium to classify the probability distributions of the fluctuations of macroscopic observables in critical nonequilibrium systems that is based on the use of symmetries, has been proposed.[36] The glass transition, where the system falls out of equilibrium, is a dynamic crossover and neither a dynamic nor a thermodynamic transition. Even if, strictly speaking, there is no phase transition, the dynamics in the *full* glassy phase shares some features with usual critical dynamics. Let us mention three properties that give support to this statement. The correlation functions, say for fixed waiting-time as a function of the total time, do not relax exponentially but decay in a much slower manner. In the free-energy landscape the dynamics at long-times can be associated to a wandering along flat directions (times are taken to be long but not so long as to allow for jumps over barriers in the free-energy landscape that scale with the size of the system). This property has been made precise for the solvable models mentioned above[24, 37, 38] and it has been shown under certain assumptions for finite dimensional disordered models.[28] A dynamic correlation length is expected to reach a power law dependence at sufficiently long times belonging to the same regime[21] (see also[39]). Thus, adapting arguments usually used in the study of critical phenomena, one can expect that the form of the probability distribution functions of fluctuating dynamical quantities will be constrained by the symmetries in the problem. On the experimental and numerical side, these distributions can be measured and classified. Clearly, noise measurements play a very important role in this respect. On the theoretical side, one can construct effective sigma models for the fluctuating quantities, derive from them scaling laws for the fluctuations[20, 21, 28] and, in certain cases, the actual form of the scaling functions.[22] This is the route we have followed recently.

In this presentation I review several aspects of the non-equilibrium dynamics of glassy systems, including aging relaxational phenomena and rejuvenation caused by external perturbations. Until recently, experiments, numerical studies and theory have focused on the behavior of averaged bulk quantities. The interest is now shifting to the study of dynamic heterogeneities, and intermittency. The aim is to detect similarities and differences in the behavior of temporal and spatial fluctuations in different glassy systems. I present a short summary of the clearest experimental results concerning fluctuations and I summarize the theoretical approach that we developed to describe them.[20–22, 28]

2. NONEQUILIBRIUM RELAXATION IN GLASSY SYSTEMS

Out of equilibrium relaxational dynamics occurs, for instance, when one suddenly quenches a system with ferromagnetic interactions below its Curie temperature. The system evolves from the very disordered initial condition via the growth of domains of up and down magnetic order. Since the typical domain radius behaves as $t^{1/2}$ the time needed to order a sample of linear size L goes as L^2 and cannot be reached with finite times with respect to L. When quenched disorder, as random fields, are also present the dynamics keeps the same characteristics but it is much slower.[40]

In the domain growth example one easily visualizes the growth of order. Going across the super-cooled liquid–glass crossover thorough any path (annealing in temperature, crunch in pressure, etc.) the systems fall out of equilibrium as demonstrated by several facts: the observables depend on the preparation used, the systems age, etc. Whether there is a growing order controlling their evolution in the glassy phase is still an open question. Glasses of very different types have been identified and studied and we mention only some samples that we shall discuss in this report: dipolar glasses,[40] spin-glasses,[29, 41, 42] colloidal suspensions,[3–6, 9, 10, 30–32, 43] polymer glasses,[7, 9, 43] glycerol[8, 44] and relaxor ferroelectrics.[45]

The above examples concern systems that are not able to reach equilibrium with their environments in a reasonable time but that, let evolve on astronomical time-scales, will eventually equilibrate. A liquid can be driven to a slow out of equilibrium stationary regime by a weak shear. This is a force that does not derive from a potential and its effect cannot be described with a usual statistical mechanics approach. If the liquid is sufficiently dense, its sheared dynamics resembles the one of a purely relaxing super-cooled liquid (see Fig. 1 for the details) but it occurs out of equilibrium.[16, 30, 31, 46]

Another family of materials that have captured the attention of experimentalists and theoreticians in recent years is granular matter.[47–49] Since the potential energy needed to displace a macroscopic grain by a distance equal to its diameter, mgd, is much larger than the characteristic thermal energy, $k_B T$, thermal activation is totally irrelevant for systems made of macroscopic grains. Therefore, in the absence of external driving granular matter is blocked in metastable states. There is no statistical mechanics approach capable of describing its static behavior. Instead, when energy is pumped in the form of shearing, vibration or tapping, transitions between the otherwise metastable states occur and granular matter slowly relaxes towards configurations with higher densities. Glassy features such as hysteresis as a function of the amount of energy injected, slow dynamics, and non stationary correlations have been exhibited.

Aging means that older systems relax in a slower manner than younger ones. One defines the age of a system as the time spent in the phase under study. For instance, the age of a system that is suddenly quenched from high-T to low-T is simply the time elapsed since the quench. Colloidal suspensions at a given concentration are usually initialized by applying a strong stirring that is suddenly stopped at the initial time. The aging properties are studied by monitoring the time evolution of correlation and response functions. In the former experiments one lets the system evolve and compares its configuration at a waiting-time t_w with the one reached at a subsequent time $\tau + t_w$ (see Fig. 1). In the latter one perturbs the system at t_w with, *e.g.* a dc or an ac field, and follows the evolution of the linear response to the perturbation. These are *global or bulk* functions in the sense that they represent the dynamics of the full system, and they are averaged over different repetitions of the experiment (statistical average) and sometimes a coarse-graining in time using a short time-window around the observation instant is also implemented. In the glassy phase both correlations and responses depend on t_w in an aging manner and, within the experimentally accessible time-window, this trend does not show any tendency to stop. At temperatures that are close but above T_g, or concentrations that are close but below ϕ_g, one observes "interrupted aging" that is, a dependence on the age of the system until it reaches the equilibration time $(t_w > t_{eq})$ where the dynamics crosses over to an equilibrium one. In equilibrium correlation and response measurements are related in a system independent manner by the fluctuation-dissipation theorem. Out of equilibrium this general relation does not hold and important information can be extracted from its modifications[1] (see Sect. 4.5).

Aging has an easy interpretation within coarsening systems.[1] The motion of interfaces is usually driven by their curvature. Since domains grow, the curvature of the domain walls decreases in time and the dynamics slows down as time elapses. Comparing the configuration at t_w and at a later time $\tau + t_w$, one finds a clear separation of time-scales depending on the relative value of τ with respect to t_w. For short time differences with respect to a characteristic time $\tau_0(t_w) \equiv 1/[d_{t_w} \ln \mathcal{R}(t_w)]$, with $\mathcal{R}(t_w)$ the typical domain radius at t_w, domain walls do not move. Then, if the ratio between surface and volume of the domains vanishes in the thermodynamic limit (a hypothesis that might be violated in some systems), the overlap between the configurations at t_w and $\tau + t_w$ simply takes into account the thermal fluctuations within the domains and the correlation decays as in equilibrium from 1 at equal times to a value called $q_{ea}(T)$ [that equals $m_{eq}^2(T)$ in a ferromagnetic system] when $\tau \to \tau_0(t_w)$. For time-differences beyond $\tau_0(t_w)$, the correlations decay below $m_{eq}^2(T)$ since the interfaces move and one compares configurations with very different domain structures. As already mentioned, coarsening

occurs, for instance, in ferromagnets below T_c. The aging properties of the orientational glass $K_{1-x}Li_xTaO_3$ have also been interpreted using such a picture.[40]

In structural glasses, a pictorial explanation of aging is also possible imagining that each particle sees a cage made of its neighbors.[1] When τ is short compared to a characteristic time $\tau_0(t_w)$ each particle typically rattles within its cage and the decorrelation is only characterized by thermal fluctuations. As in the coarsening example, the correlations decay in a stationary manner from its value at equal times to $q_{ea}(T)$ in this time regime. When τ increases, the motion of the particles destroys the original cages and one sees the structural relaxation. The waiting-time dependence implies that the cages are stiffer when time evolves. The motion of a tagged particle observed with confocal microscopy demonstrated this scenario.[5]

By shearing a liquid one usually facilitates its flow and the relaxation time decreases with increasing shearing rate (shear thinning). By shearing a glass one typically introduces a characteristic time that corresponds to the longest relaxation time. Thus, aging is interrupted for longer waiting-times and the sample is rejuvenated by the external perturbation.[16, 30, 31, 46, 50] The two-step structure of the relaxation remains unaltered when a weak shear is applied but the scaling in time is, however, modified. This observation is key to the derivation of the theory of fluctuations we shall discuss in Sect. 4.

The effect of an oscillatory drive, typically used to provoke the relaxation of granular matter, can be rather different. Indeed, the study of mean-field models of granular matter indicated that aging might not be arrested by such a perturbation.[51] This is the subject of current experimental and numerical investigations.[48]

The curves in Fig. 1 can be scaled in a quite satisfactory way by using the following form:[1, 24, 25]

$$C(t, t_w) = C_{fast}(t - t_w) + C_{slow}(t, t_w) = f_{fast}\left(\frac{h_{fast}(t)}{h_{fast}(t_w)}\right) + f_{slow}\left(\frac{h_{slow}(t)}{h_{slow}(t_w)}\right) \tag{1}$$

with $h_{fast}(t) = e^{t/\tau}$ and $h_{slow}(t)$ a system-dependent monotonic function. In aging systems the first term describes the stationary approach to the plateau, the second term the t_w-dependent departure from it. In some cases, as the $3d$ EA model, one finds that $h_{slow}(t) = t$ describes rather accurately the available data. In some other aging systems like the insulating spin-glass studied by Hérisson and Ocio[29] an enhanced power law does a better job. Notably, the effect of a weak shear is to modify $h_{slow}(t)$ rendering it an exponential in such a way that the second decay becomes stationary as well. It only weakly modifies instead other important features of the relaxation as the value of the plateau or how the FDT (see Sect. 4.5) is modified. In theoretical terms, the important response of the scaling function h_{slow} to external perturbations is due to the time-reparametrization invariance of the dynamic action for the structural relaxation.[27, 28] The consequence of this symmetry as fluctuations are concerned will be explained in Sect. 4.

3. HETEROGENEOUS DYNAMICS: EXPERIMENTS AND SIMULATIONS

Two extreme interpretations of the origin of the non-exponential relaxation in super-cooled liquids are the so-called "heterogeneous" and "homogeneous" scenarios.[2] In the former, the non-exponential relaxation is supposed to be due to the exponential decay of independent cooperative regions (clusters of estimated 2-4 nm typical size) each with its own characteristic time. Note that any well-behaved relaxation function can be expressed as $R(t) = \int_0^\infty d\tau\, g(\tau)\, e^{-t/\tau}$. The very non-trivial ingredient in this scenario is that the exponentials are associated to the relaxation of individual and localized regions in the sample. In the latter scenario instead the sample behaves homogeneously and the decay is intrinsically non-exponential. Clearly, any intermediate scenario is also possible, with some regions transiently decaying exponentially, and later returning to the bulk behavior.

In the descriptions discussed in the previous paragraph the heterogeneities are supposed to have an exponential relaxation. More generally, an heterogeneity can be a region in the sample that relaxes very differently from the bulk but not necessarily with an exponential function. Clearly, the heterogeneities will not be static and they will appear and disappear as dynamic fluctuations. Thus, we shall adopt a more generic definition of dynamic heterogeneity that is "any mesoscopic region in the sample that transiently relaxes with a very different law from the one in the bulk".

The existence of dynamic heterogeneities in super-cooled liquids and glasses has been suggested on the basis of experiments performed using many different techniques.[2] Some of these methods test the heterogeneous character
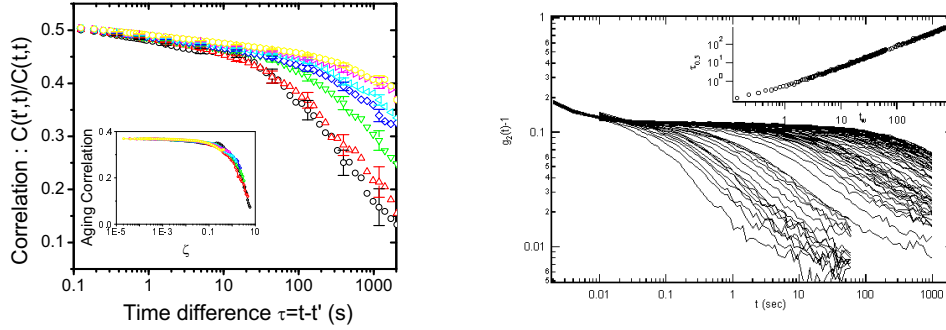
Figure 1. Left: Decay of the normalized two-time correlations between magnetic fluctuations,[29] $m_\alpha(t)$, in the insulating spin-glass $CdCr_{1.7}In_{0.3}S_4$ (the index α indicates the number of the experimental run). The magnetic fluctuations are measured using a short temporal coarse-graining. The correlation $C(t, t_w)$ is obtained by averaging over $\mathcal{N}$ repetitions of the same experience the product of m_α at two subsequent times: $C(t, t_w) \propto \mathcal{N}^{-1} \sum_{\alpha=1}^{\mathcal{N}} m_\alpha(t) m_\alpha(t_w)$. Each curve corresponds to a fixed waiting-time and it represents C as a function of the time-difference. The lowest (uppest) curve corresponds to the shortest (longest) waiting-time. Right: Decay of the normalized intensity-intensity correlation function, $g_2(t, t_w) - 1$, measured with light scattering on a colloidal system in its glassy phase.[31] The curves are averaged over different runs and using a temporal coarse-graining. The same system under a permanent shear presents very similar decorrelation with the unique difference that the second decay below the plateau is now stationary with a characteristic time determined by the shear strength.[16, 46, 50]

in a very indirect manner.[52] Others, have an easier interpretation and yield a more direct probe of the structure and dynamics. In this Section we summarize the evidence for heterogeneous dynamics in super-cooled liquids and glasses focusing only on the results obtained with light-scattering, atomic-force probes, and confocal microscopy. We also briefly review the numerical simulations done with the purpose of studying dynamic heterogeneities. Most of these works have concentrated on the super-cooled liquid phase of a binary Lennard-Jones mixture[11–16] and polymer melts[17] in two and three dimensions and the heterogeneities have been studied by tagging each particle and classifying them according to their mobility during a selected time-interval. Some recent papers treat the glassy phase of the particle system in a similar way.[19]

Following the dynamics of individual particles is very useful to get an intuitive understanding of the evolution of the system but it is hard to use as a direct input in a theoretical approach. These results, as well as the numerical analysis of spin models,[20, 21] suggest to use a coarse-grained description of the local properties. We shall discuss this approach[20, 21, 28] in Sect. 4.

3.1. Spatial heterogeneities

Israeloff and collaborators used non-contact scanning probe microscopy techniques (cantilevers)[7] to measure the fluctuations in the dielectric properties of various polymer films at a mesoscopic – nanometric – scale. Working at and below their glass transition these authors found a polarization noise power spectrum that is typically $1/f^\gamma$ but whose form or exponent vary in time. This variation was associated to transient dynamical heterogeneities with lifetime similar to the α-relaxation time. Interestingly enough, the appearance of smooth Lorentzian peaks in the spectrum was accompanied by the presence of random telegraph switching between a few discrete levels in the polarization time series. These features lasted between a few seconds and at most a few hours showing the transient but sometimes long-lived nature of the heterogeneity.

Confocal microscopy allows one reconstruct the trajectory of each particle in a three dimensional colloidal suspension made of several thousand particles.[3, 4] Using this technique Kegel and van Blaaderen[3] found a non-Gaussian distribution of particle displacements in a dense system of colloidal hard spheres and they associated this large distribution to the presence of dynamical heterogeneities.

A visual way of characterizing heterogeneities in the dynamics of a particle system is to classify the particles according to their mobility during a chosen time-interval and then study the geometric and dynamic properties

of these subensembles. Even if there is no unique definition of the mobility of a particle, several reasonable definitions have been used[13]* numerically and experimentally and particular attention has been payed to the behavior of the two extreme cases of, say, the 5% most mobile and 5% most immobile particles.

The molecular dynamic simulations of Donati el al[13] show that at any given instant most of the particles in a super-cooled liquid oscillate within their cages apart from $5\% - 6\%$ that move quickly along stringy paths on which particles follow one another. The mean cluster size of mobile particles[†] increases when lowering the temperature suggesting the existence of a percolation transition at the estimated mode-coupling temperature T_c with a divergence of the cluster size in the thermodynamic limit. The fractal dimension of these clusters is close to 1.75. In contrast, the immobile particles, i.e. those with a small self-displacement, group in rather compact clusters with a mean cluster size that is quite independent of T. The mobility of a particle is directly related to its potential energy, with the most mobile (immobile) particles having the highest (lowest) potential energy. There is also a correlation between mobility and local composition, with immobile particles being related to small composition fluctuations in the mixture. The time-dependent correlation functions, restricted to the mobile particles only, have a very similar structure to the bulk ones with a two-step decay and a cage effect, only that the life-time of the cage is shorter than on average and the second decay can be seen in time differences on which the bulk correlation only reaches the plateau. Very few particles retain their mobility after the chosen time-interval, $i.e.$ the fast or slow character of a particle is renewed. The relaxation does not correspond to "independent" volumes relaxing exponentially each with its own relaxation time.

The organization in mobile and immobile particles has also been observed in the colloidal dense liquids studied with confocal microscopy and Weeks el al showed that, indeed, the typical cluster size grows when approaching the glass transition.[4, 5]

Fewer studies of spatial heterogeneities in the glassy phase exist.[18–21] Vollmayr-Lee et al analyzed the structure of the binary Lennard-Jones mixture below its glass transition[19] using a similar approach to the one developed by Donati el al[13] for the super-cooled liquid phase. In short, they identified the most mobile and most immobile particles during a sufficiently short time-interval and at sufficiently low temperature such that aging effects can be neglected. These authors showed that mobile (and immobile) particles tend to be near each other also in the glassy phase. As expected, mobile (immobile) particles are placed in regions of lower (higher) density than average and wider (narrower) cages surround mobile (immobile) particles. The identity of mobile and immobile particles changes in time also in this phase.

Experimentally, Courtland and Weeks also found clusters of very mobile particles in the glassy phase of a colloidal system.[6] (Note that a short coarse-graining in time was implemented in these measurements in contrast with the previous studies in glycerol where clustering was not observed.[8]) Interestingly enough, no evidence for a change in the properties of these clusters as the sample ages has been reported, suggesting that clusters of very mobile particles exist even in a very old sample.

The behavior described above can be confronted to the geometric structure of clusters of spins with similar values of the coarse-grained two-time correlations in the aging dynamics of a spin-glass where quenched disorder is very important in determining the spatial properties of each sample.[21] We found that clusters of negatively valued coarse-grained two-time correlations that correspond to very fast, under constrained spins, are well localized. In contrast, clusters of positively valued, slower spins, are practically space-filling.

3.2. Intermittency

Large fluctuations in the time series of a global quantity can be associated to sudden large rearrangements occurring somewhere in the sample. Thus, the study of fluctuations in the instantaneous measurement of a global quantity can be very useful to characterize the spatial dynamics.

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*Donati et al chose the time Δt over which the displacement $|\vec{r}_i(t + \Delta t) - \vec{r}_i(t)|$ is calculated to be the one that yields the maximum non-Gaussianity of the – spatial – van-Hove correlator.[13] This time lies between the end of the β-relaxation and the beginning of the α-relaxation The mobility of a particle is then defined as its maximum displacement during Δt.

†Clusters of particles with a chosen mobility are usually defined as the ensemble of such particles which are closer than the first minimum of the pair static correlation function.

The nature of temporal fluctuations are usually studied via the frequency dependence of the noise spectra[41, 42, 45, 49] (search for $1/f$ noise). This quantity, being the Fourier transform of a temporal correlation function, only gives information about the second moment of the fluctuating quantity. Richer information can be extracted from the full probability distribution. Non-Gaussian distributions of many interesting observables in glassy systems have been recently reported. In the rest of this Section we define the measuring procedure and we recall these experimental results.

In a typical (numerical or real) experiment one monitors a time-dependent global observable, O, and collects the values it takes with a chosen sampling frequency. A statistical measure involves many repetitions of the experiment done in identical conditions. Usually, the reading of the observable on the time grid, $t_k = k\Delta$, is not strictly local in time but involves a temporal coarse-graining. In other words, the observable measured on run α at time t_k is the result of an average over a time window around t_k: $\overline{O}^\alpha(t_k) \equiv n^{-1} \sum_{m=0}^{n-1} O^\alpha(t_{k-1} + m\delta)$ with $n\delta = \Delta$. The coarse-graining in time should not significantly change the main properties of the time-series. However, it is clear that the longer the time-interval Δ, keeping δ fixed, the smoother the signal. Using an excessive averaging might erase the effect of rare events and hide the intermittent character of the relaxation.

Given the time-series for $\overline{O}^\alpha(t_k)$ one can then check how the comparison of the signal taken at two-times separated by a constant lag τ evolves in time. That is to say, one takes $C^\alpha(t_k + \tau, t_k) \equiv \overline{O}^\alpha(t_k)\overline{O}^\alpha(t_k + \tau)$ and follows its evolution as t_k spans the interval $[t_1, t_2]$.[‡] (In Fig. 1 this procedure corresponds to moving vertically at a constant value τ of the temporal axis.) Choosing t_1 very long one ensures that the averaged bulk correlations, and possibly the result of each run, decay very slowly in two steps. If τ does not go very far beyond t_1, for all t_k one explores times and time-differences that fall on the β-relaxation. With the collection of data points $C^\alpha(t_k + \tau, t_k)$, for α ranging from 1 to the number of runs, and $k = 1, \ldots, (t_2 - t_1)/\delta$ one can construct a histogram or a probability distribution. It turns out that its form and its dependence on the times t_1 and τ, yield very interesting information about the dynamics of the samples.

The "time-resolved light-scattering technique"[10] has been developed with the aim of testing these fluctuations. Using a multi-speckle collector, $C^\alpha(t_k + \tau, t_k)$ is calculated as an average over speckles of the intensity-intensity two-time correlations in light scattering measurements. Temporal fluctuations in colloidal suspensions have been studied in this way. Interestingly enough, this group has shown that aging samples, such as a concentrated colloidal gel, have an intermittent dynamics leading to negatively skewed distributions of the two-time intensity-intensity correlations. This non-trivial temporal behaviour, once averaged over a short time-window, lead to a decay of the intensity correlation function, $g_2 - 1$, as the one shown in the right panel of Fig. 1.

Evidence for intermittency in the *global* voltage signal noise in laponite, a solid-like colloidal glass, and Makrofol DE 1-1 C, a polymer glass, has been presented recently.[9] While the samples are out of equilibrium and aging, as proven by independent light scattering measurements in the case of laponite[30] or the non-stationarity of the noise spectrum in the case of the polymer glass, the voltage time series has bursts with very large amplitude. With sample age these bursts become rarer in the sense that their amplitude decreases and the time elapsed between two consecutive ones increases. Eventually, when aging stops the bursts disappear from the noise signal. In order to better characterize the signal Buisson *et al*[9] constructed a waiting-time dependent PDF, $\rho(V)$, by dividing time in intervals of duration τ. The PDFs are clearly negatively skewed (intermittent noise) at short times and they progressively approach a Gaussian distribution when the system approaches equilibrium.

Using confocal microscopy Courtland and Weeks demonstrated that the averaged (over all particles) displacement between a waiting-time and a subsequent time is also intermittent leading to a non-Gaussian distribution.[6] It is interesting to note that these experiments show that significant fluctuations occur even if the time-lag τ is very short compared to t_w.

4. HETEROGENEOUS AGING DYNAMICS: THEORY

Recently, we constructed a framework for the study of fluctuations in the nonequilibrium relaxation of glassy systems with and without quenched disorder.[20–22, 28] In these articles we studied coarse-grained local correlators obtained for a given noise or experimental realization, and the fluctuations of two-time global quantities in finite-size systems, but we did not study the temporal fluctuations, leading to intermittent or continuous dynamics as

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[‡]Note that this quantity is not a correlation function since there is no averaging in its definition.

found experimentally.[9, 10] Here we present preliminary results concerning the behavior of the latter fluctuations in spin-glass models.[22] Focusing on the fluctuations of the coarse-grained local two-time quantities we predicted constraints on their distributions.[20–22, 28] In particular we showed that locally defined correlations and responses are connected by a generalized local out-of-equilibrium fluctuation-dissipation relation. We argued that large-size heterogeneities in the age of the system should survive in the long-time limit. A symmetry of the underlying theory, namely invariance under reparametrizations of the time coordinates, is at the basis of these results. We established a connection between the probabilities of spatial distributions of local coarse-grained quantities and the theory of dynamic random manifolds. We defined and discussed the behavior of a two-time dependent correlation length from the spatial decay of the fluctuations in the two-time local functions. For concreteness, we presented numerical tests performed on disordered spin models in finite and infinite dimensions. We characterize the fluctuations in the system in terms of clusters of coarse-grained sites with similar properties, similarly to what has been done when studying super-cooled liquids and glasses numerically[13] and experimentally.[4–6] Finally, we explained how these ideas can be applied to the analysis of the dynamics of other glassy systems that can be either spin models without disorder or atomic and molecular glassy systems. In this Section we review part of these results and we present some more recent studies of the form of the probability distribution functions of the two-time correlation functions.

4.1. Distinction between fluctuations: spatial, dynamical and finite size

Let us define several two-time auto-correlations that differ in how they have been averaged. For concreteness, we consider a cubic system with fixed volume $V = L^d$.

4.1.1. Temporal fluctuations in global measurements

In numerical simulations of Ising spin models a useful choice for the observable is $O^\alpha = N^{-1/2} \sum_{i=1}^N s_i^\alpha$. Note that $C^\alpha(t_k + \tau, t_k) = N^{-1} \sum_i s_i^\alpha(t_k + \tau)s_i^\alpha(t_k)$ (since crossed terms cancel) and it is normalized to one at equal times. Interestingly enough, the PDF of C^α takes a very similar form[22] to the one found in the light scattering measurements of some colloidal suspensions[10] (see Fig. 2). For infinitesimal short lag-times τ one expects a Gaussian distribution of fluctuations but this regime is not of easy access neither numerically nor experimentally. For longer but still short lags that correspond to the end of the β-relaxation one finds a negatively skewed distribution with a rather long tail towards small values of C. For much longer lags such that one enters deeply in the structural relaxation regime the distribution gets more symmetric and eventually becomes Gaussian again.[10, 22]

The (two-time dependent) negatively skewed PDF can be rather well described with a *generalized Gumbel distribution*:[10, 22]

$$\rho_b(x) = \frac{|b|}{\Gamma(b)} e^{b \ln b} e^{b\left(\alpha(x-x_o) - e^{\alpha(x-x_o)}\right)} . \tag{2}$$

When $b = 1$ this is the usual Gumbel distribution of the first kind. When $b = \pi/2$ Bramwell, Holdsworth and Pinton[54] found that it describes very accurately the fluctuations in the energy injected in a close turbulent flow at fixed Reynolds number, it characterizes exactly the fluctuations in the $2d$ XY model close to its critical line and it also describes many other types of fluctuations in very different critical systems. Rácz *et al* discussed these distributions in connection with the roughness of random surfaces and $1/f$ noise.[36, 53] In Sect. 4.3 we shall discuss a justification to use this distribution based on an effective random manifold model that we proposed controls the fluctuations in the temporal and spatial correlations.[20–22, 28] We shall argue that it might be possible to scale the PDF's for all t_w and t using modified Gumbel distributions as in (2) with a parameter b that is real and smoothly two-time dependent for aging problems.

4.1.2. Spatial fluctuations

In Sect. 3 we described the observation of local dynamic heterogeneities in some glassy systems. These studies focused on single particle measurements. In order to derive a theoretical model including these effects it is more convenient to introduce a spatially coarse-grained description. With this aim let us divide the system with volume $V = L^d$ into n subsystems with volume $v = \ell^d$. One can then compare the configuration of the system within the same local volume at two different times. This defines what we call a two-time coarse-grained local
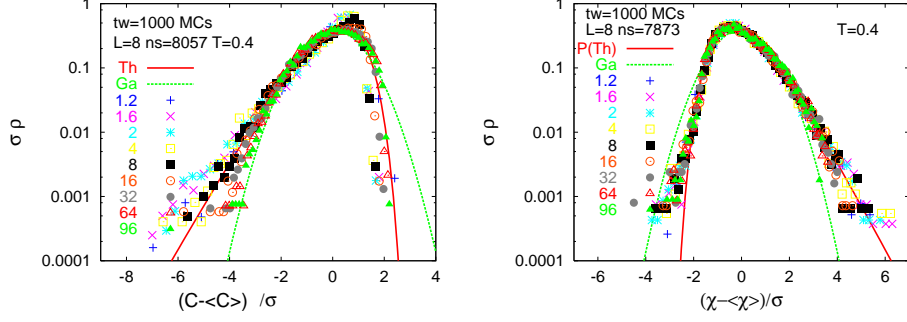
Figure 2. Scaled probability distribution of the mesoscopic fluctuations in the global correlation, $N^{-1}\sum_{i=1}^{N} s_i(t)s_i(t_w)$ (left), and susceptibility, $N^{-1}\sum_{i=1}^{N}\delta s_i(t)/\epsilon_i(t_w)|_{\epsilon=0}$ (right), of the $3d$EA model with linear size $L=8$ at $T=0.4 < T_c$ obtained form Montecarlo simulations.[22] σ and $\langle C \rangle$ (and $\langle \chi \rangle$) are the variance and average of the raw PDF, ρ, at each pair of times. A temporal coarse-graining of the spin configuration has been implemented, with a coarse-graining time equal to $t/10$. Different sets of points correspond to different values of t/t_w that are given in the key. The waiting-time is $t_w = 10^4$ MCs. n_s is the number of runs, each one leading to a point in the generation of the histogram. The red line is a generalized Gumbel PDF[54] with $b = \pi/2$, see Eq. (2). The green line is a Gaussian PDF with the same variance. See Ref.[22] for more details on the functional form of these PDFs.

correlation (even if, strictly, this is not a correlation). Again, this is a single run quantity. To obtain better probability distributions one can repeat the measurement and get L/ℓ data points for each run.

For an Ising spin model on a lattice we have $C_i^{cg}(t, t_w) \equiv v^{-1}\sum_{j \in v_i} s_j(t)s_j(t_w)$, where v_i is the coarse-graining volume surrounding the site i that equals the number of spins within this volume. For simplicity, one takes it to be the same for all sites in the sample. We omitted the index α labelling runs to make the notation more compact. The local coarse-grained correlation is normalized to one at equal times. Another interesting example is the case of a particle system in a d dimensional continuum space. In this case, the density fluctuations are probably the simplest and most interesting observables. One then has $C_i^{cg}(t, t_w) \equiv (\rho_i(t) - \langle\rho(t)\rangle)(\rho_i(t_w) - \langle\rho(t_w)\rangle)$ that one can normalize to be equal to one at equal times.[57]

If we use a sufficiently large coarse-graining volume v, the local correlations should maintain the more salient qualitative features of the decay of their bulk partners.[20, 21] Thus, the local correlations should decay quickly towards a *local* Edwards-Anderson parameter, $q_{ea}^i \equiv \lim_{t-t_w \to \infty} \lim_{t_w \to \infty} C_i(t, t_w)$, and then slowly below this value. If the structure of the global correlation is preserved at the local level, the first step of the relaxation should be stationary whilst the second one could be waiting-time dependent. Even if site-to-site fluctuations in q_{ea}^i are in principle possible, we expect them to be erased by using a sufficiently large coarse-graining volume v, and have $q_{ea}^i = q_{ea}$. The decay below the site-independent plateau should then carry all the information about the spatial fluctuations. Extending the scaling of monotonically decaying two-time correlations within a correlation-scale,[25] and using the fact that we expect that the effective dynamic action describing the dynamics for well-separated times in the structural relaxation regime becomes time-reparametrization invariant,[28] we argued that the local coarse-grained correlations in the slow regime should scale as[21]

$$C_{slow}^i(t, t_w) = f_{slow}\left(\frac{h_{slow}^i(t)}{h_{slow}^i(t_w)}\right) = f_{slow}\left(e^{\phi_{slow}^i(t)-\phi_{slow}^i(t_w)}\right) . \tag{3}$$

for all sites in the sample. In what follows we omit the subindex *slow* since we always refer to this regime. This proposal implies that different sites can evolve on totally different time-scales or there can be small local fluctuations in the scaling function $h_i(t)$ with respect to the global scaling function $h(t)$, see Eq. (1), such that $h_i(t) = h(t) + \delta h_i(t)$, with $|\delta h_i(t)| \ll h(t)$. We expect the latter behavior to be more common since rapid variations in C_i – in space and time – should be unfavored by some stiffness making these variations costly. Note that the value of the correlations on two sites i, j may cross each other as a function of time though we expect this to happen at very short time-differences only.

4.1.3. Finite size fluctuations

Another interesting kind of fluctuations are those associated to the finite (say mesoscopic) size of a sample. The idea is, simply, to study the fluctuations in the reading of a global quantity between different repetitions of the same experiment.

4.2. Sigma model

We recently studied the symmetry properties of the dynamic action for the aging dynamics of d-dimensional spin-glasses.[28] We derived a disorder-averaged dynamic generating functional that is a path integral over local coarse-grained two-time functions. The aim of this work was to show that for longer and longer waiting-times the action for the slow parts of the two-point functions that describe the structural relaxation progressively acquires a global symmetry (a zero mode develops) that is the invariance under identical transformations of time $t \to h(t)$ for all local functions. (In this proof we assumed that there is a *local* separation of time scales and that the system is causal.) Note that this means that the effective slow system becomes critical and the arguments discussed in the Introduction apply. The contributions to this action having an origin in the fast part of the relaxation (the approach to the plateau) act as a weak symmetry breaking field that selects a scaling function $h(t)$ that determines the dynamics of the global two-time functions. Now, fluctuations can be of two types. Either the external function f (longitudinal) or the internal function h (transverse) fluctuate from site to site. The symmetry of the action indicates that the former are less favorable while the latter cost the lowest action. Thus, the more favorable fluctuations correspond to spatially-varying time reparametrizations, $h(\vec{r}, t) = h(t) + \delta h(\vec{r}, t)$. We expect that a stiffness will be generated once the two-time quantities are coarse-grained, making sharp variations in $\delta h(\vec{r}, t)$ difficult to achieve. One of the main consequences of this analysis is that the coarse-grained two-time correlations and susceptibilities should behave as described in Sect. 4.5.

4.3. Effective random surface theory

Unfortunately, we are not able to study the exact dynamic generating function for a glassy problem in finite dimensions. Still, guided by symmetry constraints we can propose a phenomenological effective action for the slow modes and derive generic conclusions from it.[20–22, 28] Indeed, we expect the actual effective action for the slow modes to become invariant invariant under global time reparametrizations in the long waiting-time limit. Therefore, any phenomenological effective action we can propose must have this symmetry. We then select from all possible time-reparametrization invariant terms the ones that we estimate are the leading one in the long times limit. These include a term penalizing rapid variations (in space and time) of the coarse-grained local two-time functions that involves a gradient and time-derivatives and other "potential" terms that are dictated by the interactions in the system. The saddle-point is time-dependent but uniform in space so it is completely determined by the second type of terms. It fixes, for example, the function f and the global scaling function h in Eq. (1). The gradient term is very important to determine the fluctuations. Defining a proper time $\mathcal{T} = \ln h(t)$, and parametrising the fluctuations with the fields $\phi(\vec{r}, \mathcal{T})$, we proposed[20–22] that the action controlling the fluctuations in generic systems with slow dynamics is a random surface theory in d dimensions. This proposal allowed us to derive the time-scalings of the PDF's of local correlations[20] and we recently exploited it[22] to justify the Gumbel-like form of the PDF's, see Fig. 2.

4.4. Two-time dependent correlation length

The fluctuating quantities in the theory are two-time "fields" associated to the coarse-grained local correlations and responses. In the asymptotic limit, the time-reparametrization invariance implies a true Goldstone, or zero mass, mode. Therefore, the spatial correlations in the fluctuations should show a power law decay $\sim 1/r$ in $3d$. However, for any finite time, this symmetry is explicitly broken by irrelevant terms that play the role of symmetry breaking fields and the Goldstone mode has a small mass. In a simulation we expect to find a finite correlation length for the the coarse-grained local two-time quantities. A convenient measure of the spatial correlations in the fluctuations is

$$B(r; t, t_w) \equiv \frac{\tilde{A}(r; t, t_w) - \tilde{A}_\infty(t, t_w)}{\tilde{A}(r = 0; t, t_w) - \tilde{A}_\infty(t, t_w)} \qquad \text{with} \qquad \tilde{A}(r; t, t_w) \equiv \left[\frac{1}{N} \sum_i C_{\vec{r}_i}(t, t_w) \, C_{\vec{r}_i + \vec{r}}(t, t_w) \right] \qquad (4)$$

Fixing the relation between t and t_w to have, say, a given global correlation, one should find a correlation length $\xi(t, t_w)$ that increases monotonically for increasing t_w. We have recently studied this quantity for a finite d spin-glass model[21] and found that $\xi(t, t_w)$ takes very small values for the times reachable numerically showing that the system is very far from its true asymptotic regime. We refer the reader to this reference for more details on this quantity.

4.5. Fluctuations in the local effective temperatures

The analytic solution to mean-field disordered spin models[1, 24, 25] includes rather simple modifications of the equilibrium FDT relations between induced and spontaneous fluctuations. This new relation reads

$$\lim_{\substack{t_w \to \infty \\ C(t, t_w) = \mathtt{C}}} \chi(t, t_w) = \tilde{\chi}(\mathtt{C}) , \qquad (5)$$

where $\chi(t, t_w)$ is a linear response function that has been integrated over time from t_w to t and C is its conjugated correlation. $\tilde{\chi}$ is a model-dependent function that relates this two-time quantities. One finds that for glassy systems, $\tilde{\chi}(C) = 1/T(1 - C)$, with T the temperature of the environment, for values of the correlations that are above the plateau, and $\tilde{\chi}(C)$ takes a different linear form for values of the correlation below the plateau. For theoretical spin-glass models the second part of the function is curved. This phenomenon has been tested numerically[1] and experimentally[9, 29, 43, 44] in a very large variety of systems with slow dynamics.

Let us now define a generalized *local* fluctuation-dissipation relation (FDR) via the limit[21]

$$\lim_{\substack{t_w \to \infty \\ C_i(t, t_w) = \mathtt{C}_i}} \chi_i(t, t_w) = \tilde{\chi}_i(\mathtt{C}_i) \qquad (6)$$

that exists if the local correlations are monotonic functions of t for fixed t_w. Based on thermometric arguments,[55] we associate the variation of $\tilde{\chi}_i$ with respect to C_i to a local effective-temperature. The question now arises as to whether the fluctuations in C_i and χ_i, and the local effective-temperatures, are independent or whether they satisfy certain relations.

In the $C - T\chi$ plane, the soft (massive) modes discussed in Sect. 4.2 correspond to displacements along (transverse to) the global $\chi(C)$ curve. Thus, the spatial fluctuations in the FDR should be such that the projection of the joint PDF, $\rho(C_i, \chi_i)$, computed at two fixed times t and t_w concentrates along the global $\chi(C)$ curve. In Fig. 3 we show a sketch of this quantity. Given a pair of times $t_w \leq t$, we depict the N points $(C_i(t, t_w), T\chi_i(t, t_w))$ with arrows that represent the velocity of the points [*i.e*, the rate at which the $(C_i, T\chi_i)$ positions change as one changes t], and are located at their position in the $C - T\chi$ plane. In the same plots we draw the parametric plot for the global $T\tilde{\chi}(C)$ constructed as usual[25]: for a fixed t_w we follow the evolution of the pairs $(C, T\chi)$ as time t evolves from $t = t_w$ to $t \to \infty$. We scale the y-axis by temperature to work with dimensionless variables.

The scaling in Eq. (3) means that different sites might evolve on different time-scales and hence have their own effective temperature. Based on the analytic study of the fluctuations in a disordered spin model[28] we expect the local responses and correlations to be constrained to follow the global curve, *i.e.*

$$\tilde{\chi}_i(C_i) = \tilde{\chi}(C_i) , \qquad \text{and} \qquad -\beta_i^{eff}(C_i) \equiv \frac{d\tilde{\chi}_i(C_i)}{dC_i} = \frac{d\tilde{\chi}(C_i)}{dC_i} = \frac{d\tilde{\chi}(C)}{dC}\bigg|_{C=C_i} . \qquad (7)$$

If there are only two time-scales for the decay of the global correlation, below q_{ea} $\tilde{\chi}(C)$ is linearly dependent on C and this equation yields $\beta_i^{eff} = \beta^{eff}$, for all sites, and values of $C_i < q_{ea}$. If, on the contrary, the global correlation decays in a sequence of scales and $\tilde{\chi}(C)$ is not a linear function of C, one has fluctuations in the local effective temperature due to the fluctuations in C_i. This behavior is sketched in Fig. 3. for a system with two global correlation-scales (left) and a system with a sequence of global correlation-scales (right).

If the two times t and t_w are close to each other, in such a way that the global correlation between them lies above q_{ea}, the global correlator and the global linear integrated response are stationary and related by the FDT. In this regime of times we also expect the local quantities to be linked by the FDT. Note that the arguments based on time reparametrization invariance do not apply to these short time-differences since the relaxation is not slow here. We have verified numerically[20] that the fluctuations of the local coarse-grained correlations and responses are concentrated rather spherically around the global value $\chi = (1 - C)/T$ when $C > q_{ea}$.
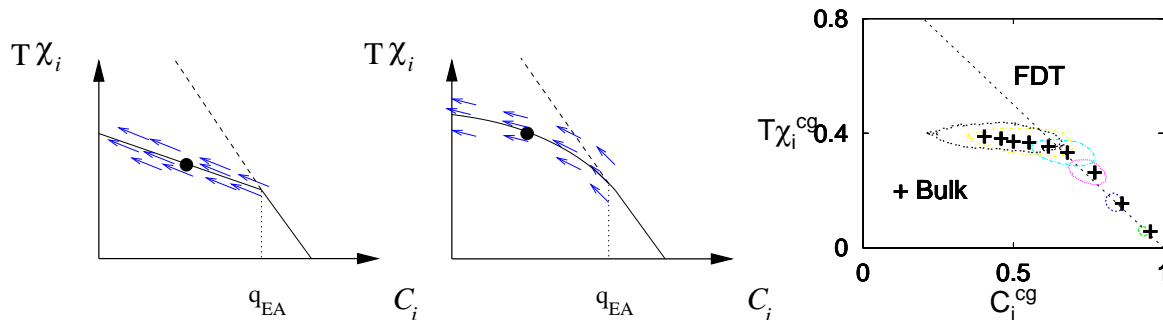
Figure 3. Left: Sketch of the fluctuations in a system with a single correlation scale below q_{ea}. The symmetry argument implies that all pairs are concentrated along the global straight line with little dispersion perpendicular to it. The arrows have the direction of the second slope in the global $\tilde{\chi}(C)$ curve. Center: A system with a sequence of correlation scales below the global q_{ea}. All point positions are constrained to be near the global $\tilde{\chi}(C)$ curve for $C < q_{ea}$, and their velocities are forced to be parallel to the global $\tilde{\chi}(C)$ curve for $C < q_{ea}$. The velocities fluctuate from site to site, but since they are determined by the global curve, they are identical for all sites with the same value of the local correlation C_i. In both cases the dashed line continues the FDT line and the black dot indicates the location of the values for the susceptibility and correlation averaged over the distribution. Right: results from a Montecarlo simulation of the $3d$ EA model below its critical temperature,[20, 21] the coarse-graining volume is $v = \ell^3 = 13^3$ and $L = 32$. The crosses correspond to the averaged values and the lines are the projection of one selected contour level for the same pair of times.

5. CONCLUSIONS

Experimental, numerical and theoretical effort is presently devoted to elucidate the role played by fluctuations in the dynamics of super-cooled liquids and glasses. From our point of view[20–22, 28] this problem can be tackled theoretically by using effective sigma-models constrained by the symmetries in the problem and, most importantly, by the expected asymptotic time-reparametrization invariance. This approach allowed us to make precise predictions on the form of the individual and joint probability distribution functions that can be put to numerical and experimental tests. The results of such measurements will yield very useful feedback to fix the terms in these effective models. Presumably, some kind of "universality" will apply and one can expect to find that a sigma-model represents the behaviour of many different classes of glasses.

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